Magnetic-Field-Induced Mott Transition in a Quasi-Two-Dimensional Organic Conductor

F. Kagawa,[1] T. Itou,[2] K. Miyagawa,[1,2] and K. Kanoda[1,2]

[1] Department of Applied Physics, University of Tokyo,
Bunkyo-ku, Tokyo 113-8656, Japan

[2] CREST, Japan Science and Technology Corporation,
Kawaguchi 332-0012, Japan

We investigated the effect of magnetic field on the highly correlated metal near the Mott transition in the quasi-two-dimensional layered organic conductor, κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl, by the resistance measurements under control of temperature, pressure, and magnetic field. It was demonstrated that the marginal metallic phase near the Mott transition is susceptible to the field-induced localization transition of the first order, as was predicted theoretically. The thermodynamic consideration of the present results gives a conceptual pressure-field phase diagram of the Mott transition at low temperatures.

Through extensive experimental and theoretical studies, it has been well recognized that almost localized Fermions (ALF) is the source of various interesting phenomena such as high-T_C superconductivity (SC) and colossal magnetoresistance. However, comprehension of the nature of ALF is still in progress, and many problems are left unsolved. One of them is the effect of magnetic field on ALF. In the theoretical approach to this problem, the Hubbard model with half-filled band has been often employed and attacked using Gutzwiller approximation earlier [1] and dynamical mean-field theory recently [2, 3]. The two approaches share an important prediction that ALF undergoes first-order localization transition by magnetic field, although different in detail. One example of the experimental realization in this context is liquid ^{3}He, which is often viewed as the strongly correlated liquid near the Mott localization. However, the field-induced localization transition was not observed in liquid ^{3}He up to 200 T [4]. It is argued that the discrepancy between the experimental and theoretical results is attributable to inadequacy of the lattice description of liquid ^{3}He through the Hubbard model with half filling [3].

The highly correlated electronic system with half-filled band provides another experimental stage suitable for the study of this issue. Recently, it has been established that the quasi-two-dimensional (quasi-2D) layered organic conductor, κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl (denoted by κ-Cl hereafter), is a prototype of the bandwidth-controlled Mott transition system with a half-filled single band [5, 6, 7, 8, 9]. The pressure-temperature (P-T) phase diagram of κ-Cl under a zero field is shown in Fig. 1(a) which is taken from our previous work [9]. Under pressure, κ-Cl shows the first-order Mott transition from the paramagnetic insulator (PI) to the paramagnetic metal (PM) with a critical endpoint at ~ 38 K. Thus the present Mott transition is viewed as a genuine one, which is driven only by electron-electron correlation and doesn't accompany symmetry breaking, as posutu-lated by Mott [10].

The purpose of the present work is to investigate the effect of magnetic field on the highly correlated metal near the Mott transition, namely a sort of ALF, in the quasi-2D system, κ-Cl. We performed the resistivity measurements for κ-Cl under control of temperature, pressure, and magnetic field. The results evidenced that the marginal metallic phase undergoes the field-induced localization transition of the first order, as is predicted theoretically.

First, in order to establish the P-T phase diagram under a high magnetic field above the upper critical field, H_{C2}, we performed the in-plane resistance measurements at 11 T (normal to the conducting layer) under isothermal $pressure$ $sweep$ by use of He gas. The κ-Cl crystal used here is the identical sample used in our previous work [9]. The P-T phase diagram under 11 T obtained in the present experiment is shown in Fig. 1(b), where closed and open circles represent the first-order Mott transition characterized by jump in resistance and crossover points defined by a pressure giving maximum in pressure-derivative of logarithmic resistance, $|\frac{1}{R}\frac{\partial R}{\partial P}|$, respectively. The AF order lines (dotted line) in Figs. 1 (a) and (b) are depicted identically, because NMR measurements on the fully deuterated salt, κ-d$_8$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, which is just on the border of the Mott transition, indicated that the Néel temperature is field-insensitive up to 12 T at least [11].

Above ~ 13 K ($\sim T_C$) the first-order lines under the different fields are nearly the same (see Fig. 1); the pressure-induced transition above ~ 13 K under 11 T is considered to be from PI to PM as in the zero field case. However, there is small but meaningful difference in a range of ~ 13 K $< T < \sim 24$ K: the pressures of the Mott transition under 11 T are slightly higher than in the zero field case by ~ 0.2 MPa at 14.1 K and ~ 0.1 MPa at 20.1 K, although the differences are not appreciable in the scale of Fig. 1. Below 13 K, the affect of magnetic
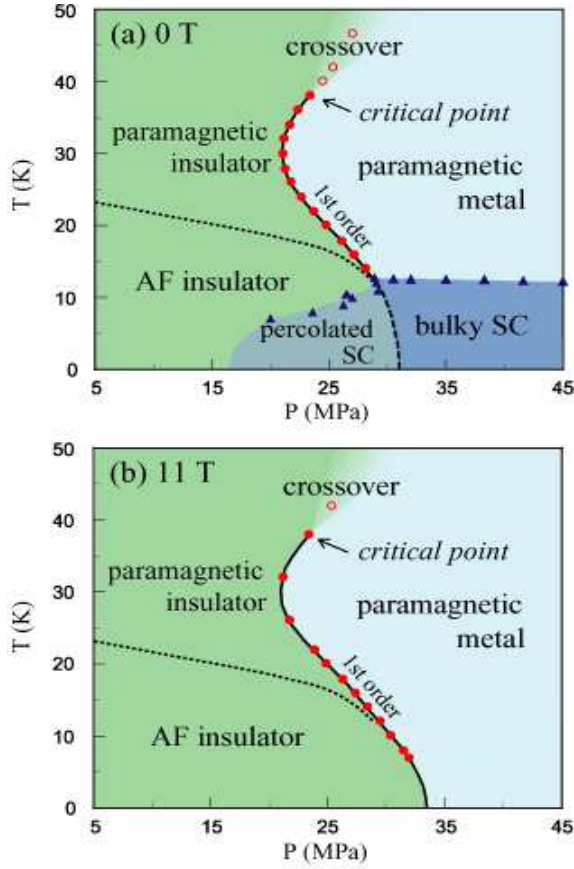
FIG. 1: Pressure-temperature phase diagrams of κ-Cl under a zero field (a) taken from Ref. [9] and under a field of 11 T normal to the conducting layer (b). Closed and open circles represent points giving the first-order Mott transition and crossover, respectively. The AF transition lines (dotted line) and the AFI-SC boundary (broken line) are taken after Lefebvre *et al.* [6].
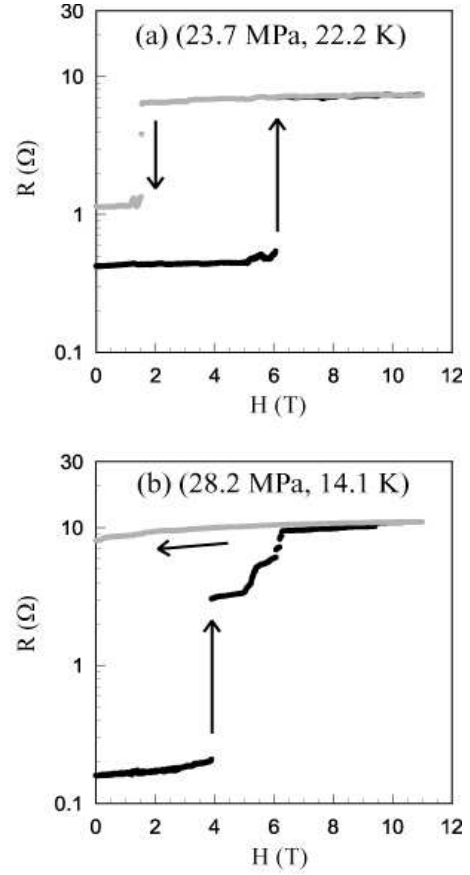

FIG. 2: Resistance curves under ascending (black) and descending (gray) fields normal to the conducting layer with temperature and pressure kept constant.

field is prominent because a magnetic field suppresses the bulky SC phase in higher pressure side and the minor SC domains [12] in the predominant antiferromagnetic insulator (AFI) in lower pressure side [9]. Thus, in the phase diagrams, percolated SC phase and bulky SC phase under a zero field are replaced by AFI phase and PM phase under 11 T, respectively. The pressure-induced transition below $\sim$ 13 K under 11 T is considered to be from AFI to PM, while it is from AFI to SC under a zero field [6]. The shift between the AFI-SC boundary [broken line in Fig. 1(a)] and the AFI-PM boundary [solid line below $\sim$ 13 K in Fig. 1(b)] is clear even in the scale of Fig. 1.

The feature that the first-order line shifts to the higher pressure side by a magnetic field implies that the marginal PM or SC undergoes the field-induced localization transition to PI or AFI. In order to demonstrate the transition, we traced the resistance behavior under *field sweep* (normal to the conducting layer) with temperature and pressure kept constant. The field sweep

was performed at an extremely slow rate ($\sim$ 0.04 T/min) after κ-Cl was adjusted at a specific point, (P^*, T^*).

In a range of $\sim$ 13 K $< T < \sim$ 24 K, we set (P^*, T^*) quite near the Mott transition boundary in PM region. The typical field dependence of the resistance is represented by the data shown in Fig. 2, which demonstrates the field-induced resistive transition of the first order clearly by the huge jump of nearly one order of magnitude and large hysteresis. The magnitude of the field of the resistance jump was very sensitive to the system position, (P^*, T^*), as expected. The P-T phase diagrams indicate that the field-induced transition at 22.2 K [Fig. 2(a)] is the localization transition from PM to PI. We emphasize that at 22.2 K a magnetic field induces a transition in the charge degree of freedom holding the spin symmetry, which is often broken concomitantly with field-induced transport transition. Thus the transition shown in Fig. 2(a) is the field-induced Mott transition without symmetry breaking. Above $\sim$ 24 K, the field-induced transition was not observed, although we chose P^* as close to the Mott transition boundary as possible with T^* fixed at 24.9 K for example [13]. This tendency is consistent with the feature of the phase diagrams that
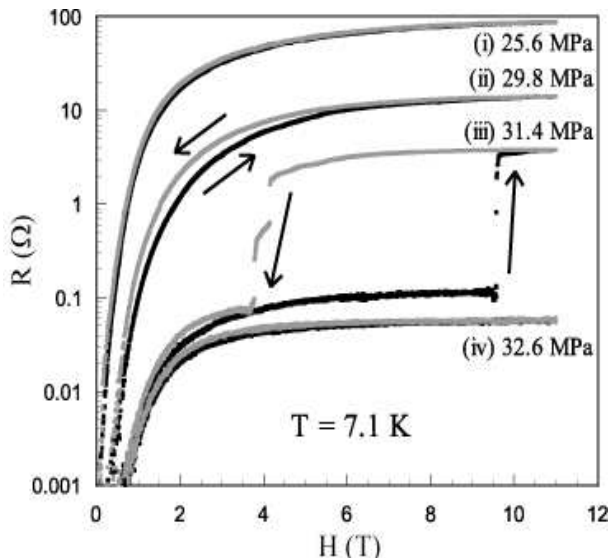
FIG. 3: Magnetic field dependence of resistance under various pressures at 7.1 K. The field is applied normal to the conducting layer The black and gray points are for ascending and descending fields, respectively.

TABLE I: Zero-field and high-field states at each pressure in Fig. 3.

(P^*, T^*)	under 0 T	under 11 T
(i) (25.6 MPa, 7.1 K)	percolated SC	AFI
(ii) (29.8 MPa, 7.1 K)	percolated SC	AFI
(iii) (31.4 MPa, 7.1 K)	bulky SC	AFI
(iv) (32.6 MPa, 7.1 K)	bulky SC	PM

transitions in Fig. 2 (above T_C) and Fig. 3 (below T_C) from the thermodynamic point of view. For clarity, we consider the pressure-field (P-H) phase diagram at a constant temperature. In the same manner of the Clausius-Clapeyron relation, the slope of the first-order line, dH/dP, in the P-H phase diagram is given as

$$\frac{dH}{dP} = \frac{V_{\mathrm{PI(AFI)}} - V_{\mathrm{PM(SC)}}}{M_{\mathrm{PI(AFI)}} - M_{\mathrm{PM(SC)}}} = \frac{\Delta V}{\Delta M}, \qquad (1)$$

where V and M indicate the system volume and magnetization respectively, and ΔV and ΔM are their jumps across the transition from PM (or SC) to PI (or AFI). The ΔV is always positive because the insulating phase is located in the lower pressure side, and is assumed to be field-insensitive up to 11 T. We first discuss the case below T_C. Since AFI in κ-Cl shows weak ferromagnetism [14] and SC shows large diamagnetism of the type-II SC under magnetic fields below H_{C2}, $\Delta M(= M_{\mathrm{AFI}} - M_{\mathrm{SC}})$ is positive and its magnitude should decrease with magnetic field increased up to H_{C2}. From Eq. (1), that leads to positive and increasing slope with a magnetic field. Above H_{C2}, ΔM is still positive because AFI phase keeps weakly ferromagnetic, and therefore the slope, dH/dP, remains positive. Thus one can draw a conceptual P-H phase diagram at $T \sim 7$ K (well below $T_C \sim 13$ K) as is shown in Fig. 4, where the solid line represents the first-order transition and the percolated SC phase is omitted for simplicity. Figure 4 indicates that magnetic field can induce either SC-AFI transition or SC-PM-AFI transition if P^* is chosen appropriate and that the Mott transition shifts to the higher pressure side with magnetic field. These features are consistent with the experimental results shown in Fig. 3 and with the shift of the first-order lines below 13 K in Fig. 1.

On the other hand, it is difficult to predict the P-H phase diagram at higher temperatures above 13 K because the sign of $\Delta M (= M_{\mathrm{PI}} - M_{\mathrm{PM}})$ in the paramagnetic region is not known. However, Fig. 2(a), which demonstrates the field-induced PM-to-PI transition, indicates that the slope, dH/dP, should be positive at least under fields of several Tesla, i.e., that PI should show larger magnetization than PM according to Eq. (1). The tendency that it is difficult to observe the field-induced transition above ~ 24 K means that the slope, dH/dP, is much steeper, i.e., that ΔM above ~ 24 K is smaller

the first-order lines have no appreciable field-dependence above ~ 24 K. Below 13 K, we performed the measurements under four pressure points at 7.1 K: (i) (25.6 MPa, 7.1 K), (ii) (29.8 MPa, 7.1 K), (iii) (31.4 MPa, 7.1 K), and (iv) (32.6 MPa, 7.1 K). The field dependence of the resistance is shown together in Fig. 3. On the basis of the P-T phase diagram, the zero-field and 11-T states at each pressure are identified as summarized in Table I (refer to this table in the following).

The SC phase away from the AFI-SC boundary [see Fig. 3(iv)] shows a typical second-order transition to PM phase. In addition to this transition, the SC phase near the boundary [see Fig. 3(iii)] shows a first-order transition with a huge hysteresis. In the ascending field process, the resistance jump is observed at ~ 9.5 T, which is far beyond H_{C2} at 7.1 K. Thus the first-order transition is the localization transition from PM to AFI. At a lower pressure of 29.8 MPa [see Fig. 3(ii)] across the AFI-SC boundary, small hysteresis without the resistance jump is observed. This implies distributed first-order transition either from non-bulky SC to AFI or through PM (see below), which likely comes from inhomogeneous internal pressure in the sample. At a further lower pressure of 25.6 MPa [see Fig. 3(i)], the magnetoresistace gets to occur at lower magnetic fields without resistance jump nor hysteresis. As is to be seen in Fig. 4 below, marginal SC remaining slightly in the host AFI by distributed internal pressure is considered to undergo SC-to-AFI transition at low magnetic fields. Invisibility of the expected hysteresis is possibly due to quite small fraction of the minor SC domains.
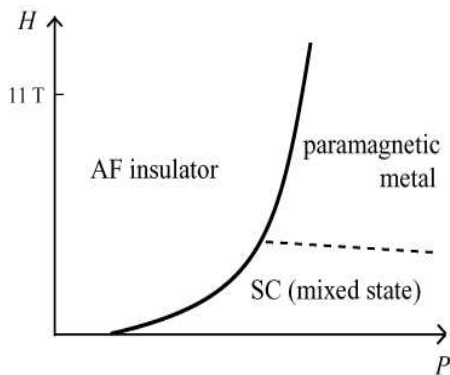
In what follows, we discuss the observed field-induced

FIG. 4: Conceptual pressure-field phase diagram at $T \sim 7$ K, which is well below $T_{\mathrm{C}} \sim 13$ K. The solid line represents the first-order transition.

than below that. Then, why does ΔM become appreciable below ~ 24 K? Two possibilities are conceivable. One, which is in an approach from high temperatures, is that the growth of ΔM reflects the establishment of Fermi liquid nature from the bad metal with temperature decreased in PM phase. As is discussed in Ref. [15], the highly correlated metal is possibly viewed as the bad metal near the critical point but as Fermi liquid at sufficiently low temperatures. According to the P-T phase diagram obtained by Limelette et $al.$ [8], the bad metal regime along the Mott transition boundary goes into Fermi liquid regime below ~ 25 K. That supports this scenario. The other possibility, which is in an approach from low temperatures, is that the large positive ΔM below T_{C} owing to the SC diamagnetization survives barely above T_{C} by the SC fluctuations. In relation to this, we note that the pseudogap is present up to two times T_{C} in the marginal PM as was evidenced by κ-d$_8$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br [16].

To conclude, we investigated the effect of magnetic field on the highly correlated metal near the Mott transition in the quasi-two-dimensional organic conductor, κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl, by resistance measurements under control of temperature, pressure, and magnetic field. The P-T phase diagram under 11 T normal to the conducting layer is established and found to have appreciable difference from the zero field case. It is demonstrated that, by application of a magnetic field, (i) the marginal PM undergoes the Mott transition of the first order and (ii) the marginal SC undergoes successive transitions: the second-order SC-PM and first-order PM-AFI ones. From the thermodynamic consideration incorporating the present results, we constructed the conceptual P-H phase diagram of the Mott transition below T_{C} in the present system. The present results evidenced the field-induced localization transition predicted from the Hubbard model, and clarified its temperature profile, which is informative for future theoretical study. On the other hand, the magnetization curve in the marginal metallic phase is an interesting future issue, because the theories predict an upward magnetization curve ($\frac{\partial^2 M}{\partial H^2} > 0$) before the field-induced transition [1, 3].

[1] D. Vollhardt, Rev. Mod. Phys. **56**, 99 (1984)
[2] For a review, see A. Georges et $al.$, Rev. Mod. Phys. **68**, 13 (1996).
[3] L. Laloux et $al.$, Phys. Rev. B **50**, 3092 (1994).
[4] S. A. J. Wiegers et $al.$, Phys. Rev. Lett. **66**, 2895 (1991).
[5] K. Kanoda, Physica C **287**, 299 (1997); Hyperfine Interact. **104**, 235 (1997).
[6] S. Lefebvre et $al.$, Phys. Rev. Lett. **85**, 5420 (2000).
[7] D. Founier et $al.$, Phys. Rev. Lett. **90**, 127 002 (2003).
[8] P. Limelette et $al.$, Phys. Rev. Lett **91**, 016 401 (2003).
[9] F. Kagawa et $al.$, Phys. Rev. B **69**, 064 511 (2004).
[10] N. F. Mott, Proc. Phys. Soc. A **62**, 416 (1949).
[11] K. Miyagawa et $al.$, unpublished.
[12] The minor SC domains near the first-order line is attributable to the distributed internal pressure of the crystal.
[13] After finishing the field sweep measurements at (22.4 MPa, 24.9 K), we depressurized the system only by 0.04 MPa, and confirmed that the resistance jump was induced.
[14] K. Miyagawa et $al.$, Phys. Rev. Lett. **75**, 1174 (1995).
[15] A. Georges, cond-mat/0403123 (2004).
[16] K. Miyagawa et $al.$, Phys. Rev. Lett. **89**, 017 003 (2002).